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ULTRASONIC RESPONSE TO TWO AND FOUR LEVEL QUANTUM SYSTEMS

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Abstract: The theory already available for the ultrasonic response to a quantum two-level system for amorphous materials is discussed and adapted to two and four level systems in crystals. It is given in a simple physical way which helps make clear the distinction between resonance and relaxation. Resonance and no relaxation occurs for small enough static strain, while for large enough strain, the opposite is true. For a TLS, relaxation occurs by a direct process, with a rate linear in temperature in the measurable range. This leads to a Debye response, but the temperature dependence is inverted from that usually found for faster relaxation rates. At low enough temperatures the relaxation strength deviates from the classical $1/T$ behavior, going to zero as the upper state becomes depopulated, while the resonance strength saturates. At high temperatures, the sum of these approaches the classical value. Relaxation and resonance strengths are given for two and four level systems as a function of temperature and strain for two strain distributions.

The theory of ultrasonic response to a two-level system (TLS) for amorphous materials taking account of distributions of internal strain and barrier heights is well developed and familiar [1,2]. Oddly enough, the theory for the simpler TLS applying to defects in crystals requiring only strain distributions seems to be less well familiar. For example, it is not generally appreciated that elastic constant relaxation due to tunneling between equivalent sites in the absence of strain (symmetric TLS) cannot occur.[3-5] The elastic constant change δC is by definition given as

$$\delta C = \partial^2 F(\epsilon) / \partial \epsilon^2 \quad (1)$$

where F is the free energy per unit volume, ϵ is an elastic strain, and C is the elastic constant belonging to that strain. Strictly speaking, C and ϵ are tensor quantities. In what follows we suppose for simplification that only one strain component is effective. The free energy is given by

$$F = -fkT \ln Z \quad (2)$$

where Z is the partition function given by

$$Z = \sum_i \exp[-(E_i/kT)] \quad (3)$$

where $E_i(\epsilon)$ are the states of the system, and f is the number of defects. There are two kinds of effects resulting for dipolar point defects. These are parelastic and dielastic [6-9], corresponding to magnetic dipolar effects. These can be illustrated with the simple two-level system shown schematically in Fig. 1. For $\epsilon = 0$, the dipole is supposed to have two equivalent orientations. The ground state is tunnel-split with a gap of $2\Delta_0$. If a stress or strain ϵ is applied, one orientation become energetically favored. For finite strain the gap 2Δ is given by

$$\Delta = (\Delta_0^2 + \alpha^2 \epsilon^2)^{1/2} \quad (4)$$

where α is a strain coupling constant. If time is available, transitions will occur, bringing the system back into thermal equilibrium.

For $T \ll \Delta$, only the ground state is populated. For small ultrasonic stress amplitudes, the elastic constant is given by the curvature of the ground state. The dipole moment is induced, proportional to the strain ϵ , and the change is called dielastic. This response has a resonance-like character. The resonance frequency is normally far above the MHz range.

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Fig. 1. Schematic illustration of potential, energy levels, transition processes, transition rates and relaxation strength Δ_R for a two level system, or a four level system including dashed lines for E . Classically $\Delta_0 = 0$, τ^{-1} is Arrhenius, and $\Delta_R \propto 1/T$

As a result no attenuation is seen, but only the low frequency change of elastic constant is found. We restrict our selves in what follows to the case of $\hbar\omega \ll \Delta_0$, where ω is the ultrasonic frequency. We call $\delta C/C$ the resonance strength Δ_g , which is then given by taking $F = -f\Delta(\epsilon)$ to obtain

$$\Delta_g = \delta C/C = -(f\alpha^2\Delta_0^2)/[C(\Delta_0^2 + \alpha^2\epsilon^2)^{3/2}] \quad (5)$$

At finite temperature, the upper level becomes populated. It has opposite curvature, leading to a reduction of Δ_g with increasing temperature.

At finite temperature phonon induced transitions become possible. For the two level system shown, however, no relaxation takes place for small amplitude ultrasonic waves about $\epsilon = 0$ because the energy of the states is flat for small ϵ , maintaining equilibrium population of the states. For $\Delta_0 = 0$, the states are given by the dashed lines. This can represent a classical Snoek system, for example Fe-C, (if one of the levels is doubly degenerate). Now if a strain is applied, equilibrium is disturbed, transitions take place, and the effect is that of a permanent dipole reorienting in a field. This is a parelastic effect, represented by a relaxation process. The low frequency (high temperature) elastic constant is given by Eqs. 1-3, and the relaxation response for all frequencies by the Debye relaxation formulas

$$(\Delta/\pi) = \Delta_R (\omega\tau)/[1 + \omega^2\tau^2], \quad \delta C/C = \Delta_R/[1 + \omega^2\tau^2] \quad (6)$$

For a classical TLS system with $\Delta_0 = 0$ and $E_1 = \pm\alpha\epsilon$, one obtains with Eqs 1-3 for $\delta C/C$ the relaxation strength Δ_R

$$\Delta_R = \delta C/C = (f\alpha^2/CkT) \operatorname{sech}^2(\alpha\epsilon/kT) \quad (7)$$

For $\epsilon = 0$, this reduces to $f\alpha^2/CkT$, the familiar classical Snoek peak result [10]. Δ_R is inverse with temperature owing to the competing thermal disordering and external strain aligning effects. Also Δ_R is proportional to the defect concentration and the square of the strain coupling constant α , given by the slope of $E(\epsilon)$. The relaxation time is Arrhenius with an activation energy U_0 given by the height of the barrier. Transitions may occur by several processes, which act in parallel. Direct transition rates are independent of temperature at the lowest temperature and occur by tunneling, and increase linearly with temperature at high temperature where they become phonon-assisted. Indirect transitions through other states may also occur, but with a higher temperature dependence. The fastest rate dominates the temperature dependence. At high temperatures, in the classical regime, transitions through states near the top of the barrier are the most effective.

The factor $\operatorname{sech}^2(\alpha\epsilon/kT)$ acts as a cut-off at low temperatures. For carbon in iron, $\alpha \sim 6\text{eV}$. Using the values $\alpha \sim 5\text{eV}$, $kT \sim 1/40\text{eV}$ at room temperature, one finds that $\operatorname{sech}^2(\alpha\epsilon/kT) = 0.98$ and 0.07 for $\epsilon = 10^{-3}$ and 10^{-2} respectively. Such cut-off effects should thus be observable even in classical systems. For a distribution of strains, a suitable average over Eq. 7 should be taken. This has been done for a gaussian distribution of internal strains for the classical $E_1 = \pm\alpha\epsilon$ by Ishiguro, et al [11] in a study of the effect of uniaxial external static stress on the attenuation in p-type silicon.

An alternative and useful way of writing $\delta C/C = \Delta_g + \Delta_R$, is, using

$$f_1 = [f \exp(-E_1/kT)]/Z, \quad (8)$$

$$\Delta_g = \langle f_1 (\partial^2 E_1 / \partial \epsilon^2) \rangle, \quad \Delta_R = \langle (\partial f_1 / \partial \epsilon) (\partial E_1 / \partial \epsilon) \rangle \quad (9)$$

This is the elastic constant change for thermal equilibrium, i.e., assuming that there is sufficient time for the concentration f_1 to adjust to the thermal equilibrium value for each strain ϵ . The first term represents the elastic constant change owing to the curvature of the states for fixed concentration, and can be identified with the resonance Δ_g . $\Delta_g = 0$ for classical systems, where E is linear in ϵ . The second term represents the change resulting from the repopulation and can therefore be identified as the relaxation strength for a Debye relaxation process.

For a quantum TLS, using $E_1 = \pm \Delta$ of Eq. 4, one obtains with Eqs. 1-3,

$$\Delta_R = (f\alpha^2/CkT)[\alpha\epsilon/\Delta]^2 \operatorname{sech}^2(\Delta/kT) \quad (10)$$

$$\Delta_S = (f\alpha^2/C\Delta_0)[\Delta_0/\Delta]^3 \tanh(\Delta/kT) \quad (11)$$

The new effects are Δ_S and the factor in square brackets in Δ_R . Figs. 2 and 3 show results for Δ_S and Δ_R separately versus inverse temperature Δ_0/kT for various values of the normalized strain $\beta = \alpha\epsilon/\Delta_0$, while Fig. 4 shows the result (dashed lines) for the total modulus change $\delta C/C$. The relaxation strengths have been normalized by $f\alpha^2/C\Delta_0$. At high temperatures $\delta C/C = f\alpha^2/CkT$, independent of Δ_0 . At low temperatures Δ_R is cut-off to zero, while Δ_S achieves its maximum value. For small strain, Δ_R goes to zero as $(\alpha\epsilon/\Delta)^2$ showing explicitly that symmetric TLS give no relaxation, while for large strain Δ_S goes to zero as $(\Delta_0/\Delta)^3$.

For a random strain distribution, Δ_R and Δ_S in Eqs. 10 and 11 may be averaged over a Lorentzian distribution [12]: $N(\epsilon) = \epsilon\omega/[\pi(\epsilon^2 + \epsilon_0^2)]$. Results for a random distribution are illustrated in Fig. 5 for $\beta = 0.5$ and 2.0 , where now $\beta = \alpha\epsilon_0/\Delta_0$. The figure shows Δ_S and the sum $\Delta_S + \Delta_R$ for two strains. Also shown is the sum for the delta function distribution of Fig. 4 for these values of strain. For the distribution, the low temperature cut-off is broadened for Δ_R and Δ_S . The dependence of $\delta C/C$ at low temperature on strain is weakened because, even for large ϵ_0 , some low strain systems are included in the distribution. This also accounts for the disappearance of a maximum in $\delta C/C$ with temperature for large strains. At high temperature, the classical result for $\delta C/C$ is obtained, and at low temperatures an analytic result for $\delta C/C$ is available which permits a discussion of the dependence of the results on the gap [13]. These curves can be used in the analysis of data to obtain all the parameters of a TLS ($\Delta_0, \alpha, \epsilon_0$) [14].

For a TLS, the one-phonon transition rate has the form

$$\tau^{-1} = A\Delta_0^2 \Delta \coth(\Delta/kT) \quad (12)$$

where A is a constant [1]. At low temperatures ($T \ll \Delta$) this depends on Δ and therefore on strain. At high temperatures ($T \gg \Delta$), $\tau^{-1} = BT$ independent of Δ and therefore of strain. Since the relaxation strength in Eq. 10 is zero for $T \ll \Delta$, it is a good approximation to take BT for τ^{-1} in Eq. 6. Using Eq. 10, one then obtains

$$\Delta/\pi = \Delta_R^*(\epsilon)/[1 + (T/T_m)^2], \quad \delta C/C = \Delta_R^*(T/T_m)/[1 + (T/T_m)^2] \quad (13)$$

where $\Delta_R^*(\epsilon) = (f\alpha^2/CkT_m)(\alpha\epsilon/\Delta)^2 \operatorname{sech}^2(\Delta/kT)$, and $T_m = \omega/B$. This is a Debye response, but the temperature dependence is inverted from the familiar form found for faster relaxation rates, with a maximum in the velocity change accompanied by a dispersion in the attenuation, all multiplied by the low temperature cut-off factor $\operatorname{sech}^2(\Delta/kT)$. Since τ is independent of strain, the response remains of Debye form when Eq. 13 is averaged over a strain distribution, with only a new $\Delta_R(\epsilon_0)$ defined. When the cut-off occurs while $\omega \gg 1$, there is a region for which the attenuation is temperature independent while the elastic constant changes linearly with temperature. This may account for some unusual behavior found for deuterium trapped at oxygen in niobium [15].

For a four-level system (FLS) with $E_{1,2} = \pm (\Delta_0^2 + \alpha^2 \epsilon^2)^{1/2}$ and $E_{3,4} = \pm \alpha\epsilon$, one finds in the same way

$$\Delta_S = \frac{1}{(1 + \beta^2)^{3/2}} \left\{ \frac{\sinh(1 + \beta^2)^{1/2} x}{\cosh \beta x + \cosh(1 + \beta^2)^{1/2} x} \right\} \quad (14)$$

$$\Delta_R = -x \left\{ \frac{[\beta/(1 + \beta^2)^{1/2}] \sinh(1 + \beta^2)^{1/2} x + \sinh \beta x}{\cosh(1 + \beta^2)^{1/2} x + \cosh \beta x} \right\}^2 + x \left\{ \frac{[\beta^2/(1 + \beta^2)] \cosh(1 + \beta^2)^{1/2} x + \cosh \beta x}{\cosh(1 + \beta^2)^{1/2} x + \cosh \beta x} \right\}$$

where $x = \Delta_0/kT$.

The total $\delta C/C = \Delta_S + \Delta_R$ is shown in Fig. 4 as the solid lines, compared to the dashed lines for a TLS with the same strains. The total FLS results are similar to those for TLS. This is to be expected since the high temperature limit is given by the classical result which is the same for both systems, and the low temperature limit is determined by the ground state, which is also the same for both systems. An important difference is that the FLS has a relaxation component even for small strains, because of the linear dependence on strain for the middle two levels. These levels, however, are not coupled by phonon transitions to other levels, so that rates to achieve equilibrium for these states could be much lower than those connecting the outer states. In this case, one might observe effectively TLS behavior at low temperature.

Fig. 2. Normalized relaxation strength as a function of normalized inverse temperature for various values of normalized strain.

Fig. 3. Normalized resonance strength as a function of normalized inverse temperature for various values of normalized strain.

Fig. 4. Equilibrium relative modulus change (sum of relaxation and resonance strengths) as a function of normalized inverse temperature for various values of normalized strain. The solid (dashed) lines are for a FLE (TLS).

Fig. 5. Normalized resonance strengths Δ_R and total strength $\Delta_R + \Delta_R$ as a function of normalized inverse temperature for two values of normalized strain. The solid curves are for a random strain distribution and the dashed curves for a delta function distribution.

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